Experimental investigation of homogeneous nucleation of water in various gases using an expansion chamber

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Abstract. Nucleation is an integral part of phase transitions and plays an important role in technology. The control of nucleation in devices such as turbines, rocket and jet engines, wind tunnels, and combustion processes is key to achieving efficient, ecologically sound operation. Our paper presents information about a modernized experimental setup used for homogeneous water nucleation research. The main goal of the study is an analysis of the influence of carrier gas on homogeneous nucleation. The experimental setup based on an expansion chamber has been used for research of homogeneous nucleation over several decades and has undergone various technical improvements. In the Institute of Thermomechanics, the setup was been completely disassembled and cleaned. Pressure transducers, laser, and the data acquisition system have been replaced with modern parts. After that, the chamber was assembled, and thoroughly tested and calibrated. Our investigations were carried with argon and nitrogen as carrier gases in a range of nucleation temperatures 220 – 260 K, pressure range 80-150 kPa, at several concentrations of water vapour. The results thus obtained are consistent with literature data. The findings suggest that this experimental approach is useful for homogeneous water nucleation research.

1 Introduction

The formation of the first microscopic fragments (small molecular clusters) of a new phase is called nucleation. In the absence of particles or walls, nucleation is homogeneous; otherwise, it is heterogeneous [7]. Nucleation is an integral part of phase transitions. Therefore, it is important for understanding many such common processes as the formation of rain, snow, crystals and smog, boiling of liquids. Playing an important role in technical processes, nucleation requires special control [1]. For example, in a nozzle flow, as well as in many other technological and environmental applications, droplet formation passes through nucleation, growth, and aging stages. The size distribution and polydispersity of the precipitate depend on the initial nucleation process. Therefore, considerable research efforts are undertaken to clarify the physicochemical properties of water. These are of interest for understanding nucleation and condensation of atmospherically relevant mixtures, e.g., in cloud formation [5]. Despite numerous studies, research of nucleation of water is an important kind of investigations to understand subtleties of the phase transitions processes. An analysis of the influence of carrier gas on homogeneous nucleation is a very important task of research in this direction. The paper presents a description of the experimental setup and the results of an experimental study of water homogeneous nucleation with argon and nitrogen as carrier gases. Our investigations were carried in a range of nucleation temperatures 220 – 260 K, initial pressure range 80 – 150 kPa, at several concentrations of water vapour.

2 Experiment

Heist and He [1] assess the various experimental devices capable of quantitative nucleation rate measurements. The most significant review over the past five years was given by Wyslouzil and J. Wölk [8]. Authors described popular research methods, standard experimental techniques and conditions to make measurements during the nucleation process.

A “space to time” experimental method was described by Peng et al. [9]. This article presented tunable diode laser absorption spectroscopy (TDLAS) and light sheet technique which were integrated in the system to monitor the unsteady condensation process in a high speed expansion flow generated by the expansion system.

A novel design of a Pulse Expansion Wave Tube (PEWT) for the study of homogeneous nucleation in mixtures of vapors and gases was presented by Campagna et al. [10].

2.1 The nucleation pulse chamber

The nucleation pulse chamber (NPC) as one of the types of Expansion Cloud Chambers was developed by Wagner and Strey in 1984 [2]. Later it has been repeatedly used to investigate homogeneous nucleation of water [3-6] and has undergone various technical improvements [1-6]. In the Institute of Thermomechanics, the setup was been completely disassembled and cleaned. MKS Baratron 615A pressure transducers were installed for measuring the pressure of water vapor and the carrier gas during the preparation of the mixture. I/O Device PCI-6115 was

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installed as the data acquisition system. The laser and vacuum system were also replaced with modern parts. After assembling the setup, all the systems were checked and calibrated.

Figure 1 shows a schematic view of this experimental arrangement. Nucleation pulse chamber can be divided into three parts: nucleation reservoir (NR), recompression reservoir (RR) and expansion reservoir (ER). The mixture is prepared in the receptacle R.-Receptacle is evacuated to about $2 \cdot 10^{-5}$ mBar. Water vapor comes from the vaporizer V through the valves NV2 and V3 up to the required pressure. After recording the water pressure, the NV2 valve is closed and a carrier gas is filled up to a certain pressure through the NV1 valve. The pressure is recorded by pressure transducers P1 and P2. The mixture passes at a constant temperature. After preparing the mixture, the process of flushing the nucleation chamber begins. This is necessary to saturate the chamber walls by the mixture, so that in the experiment the mixture does not change due to the adsorption by the walls of the chamber. For expansion, Vrec and Vexp valves are closed and the required pressures are adjusted in the RR and ER nucleation chamber reservoirs using Pb1 and Pb2 volume pumps. In the NR reservoir, the prescribed pressure is maintained by the RV valve. After setting the pressures and finishing the flushing procedure, the expansion is performed. In the process of adiabatic expansion the Vexp valve opens, and after approximately 1 ms the Vrec valve opens also. This leads to pressure and temperature drop in the NR reservoir and the conditions under which homogeneous nucleation occurs are established. A following pressure increase (about 5%) leads to quenching the further nucleation of the droplets. The formed droplets are illuminated by a laser. The intensity of the transmitted laser beam is recorded by photodiode, and the intensity of the scattered light is registered with a photomultiplier. The scattered light is normalized to the transmitted light and we get series of peaks in good agreement with Lorenz-Mie theory[11]. These peaks allow us simultaneously determine drop radius and number concentration during the growth process.

To prepare for the next expansion, valves Vexp and Vrec are closed and valves V12-V16 are opened for flushing and setting of the required pressures.

In our experiments used gases from Messer manufacturer with purity for argon 99.999% and for nitrogen 99.996%. Water was cleaned in the laboratory to a purity level of 18.2 MΩ.

This experimental setup generally operate at low total pressures and the fluids formed by nucleation and growth are often sub-cooled with respect to the solid. Also, this setup does not imply retrieving particles, or characterizing them with respect to their composition or structure *in situ*. Taking this into account, comparison with the theory is sometimes difficult[8]. Nevertheless, the positive points described in the article[8] show the exceptional advantages of the complex of equipment we use.

The innovations introduced in the experimental setup allow obtaining more accurate data and made the work more convenient, as well as speeding up the experiment.

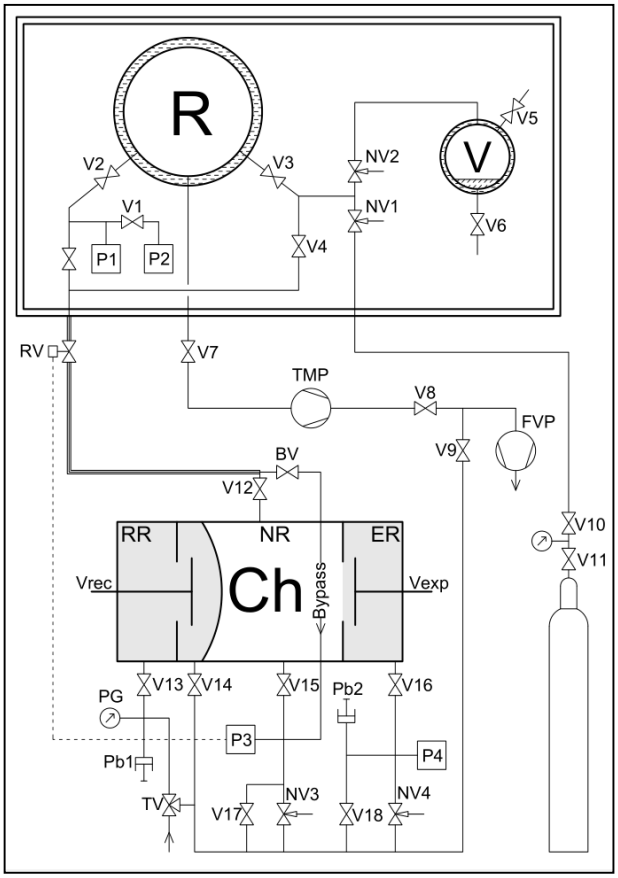

Fig.1. Schematic diagram of the homogenous nucleation setup: R – receptacle; V– vaporizer; TMP – turbomolecular pump; FVP – forevacuum pump; Ch – nucleation chamber; NR – nucleation reservoir; RR – recompression reservoir; ER – expansion reservoir; RV – electronically controlled regulation; Vx – valves; NVx – needle valves; BV - bypass valve; Vrec – recompression valve; Vexp – expansion valve; TV - three-way valve; PG – pressure gauge; Pbx – pump volume; Px – precision pressure transducers; dashed line – electric connection between regulating valve and pressure transducer in order to maintain the set pressure.

2.2 Data processing

The key parameter in quantifying the nucleation process is the nucleation rate *J* and supersaturation *S*. Nucleation rate is the number of droplets formed per unit of time and volume. For a vapor–gas mixture, supersaturation quantifies the current state deviation of the vapor

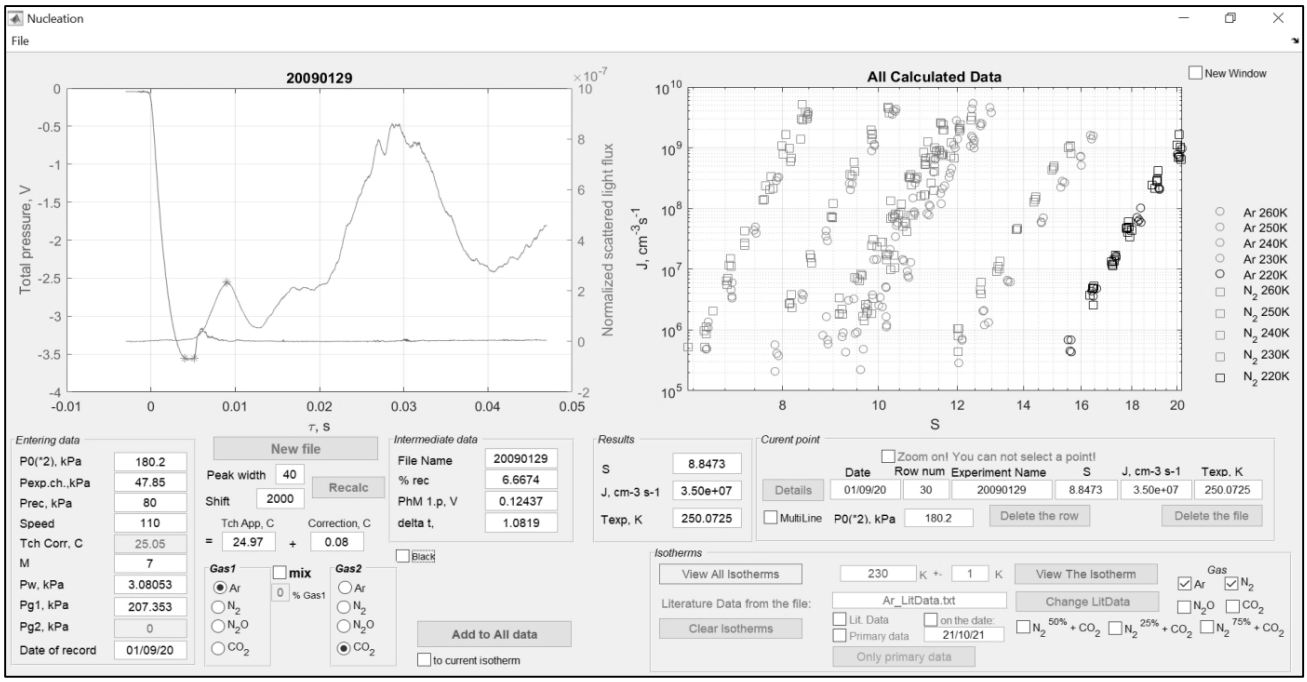

Fig. 2. The interface of the program for data processing

component from its corresponding (same p and T) phase equilibrium [10].

The classical nucleation theory (CNT) was first formulated by Volmer and Weber [12]. Later, numerous authors [13-19] improved the description, making it the most successful model for quantitatively predicting the phenomena of nucleation.

The temperature is calculated in accordance with Poisson's law

$$T_{EXP} = T_0 \left(1 - \frac{\Delta P_{EXP}}{P_0}\right)^{\frac{K_m - 1}{K_m}}, \quad (1)$$

where T_0 and P_0 initial temperature and pressure. The ratio Km of the specific heats is obtained using the Richarz formula [20].

Supersaturation is calculated by the formula

$$S = \frac{\omega P_{EXP}}{P_{ve(T)}}, \quad (2)$$

where ω is the molar fraction of the water. P_{EXP} is expansion pressure and $P_{ve}(T)$ is saturation pressure of water at nucleation temperature.

Nucleation rate is evaluated as

$$J = \frac{C_{EXP}}{\Delta t_{EXP}}, \quad (3)$$

where C_{exp} is the number concentration of droplets is observed by light scattering, as described by Wagner [21], Δt_{exp} experimental duration of the nucleation pulse.

With the help of manual settings, we can get the parameters necessary for further calculations, such as the maximum of first peak of normalized scattered light, duration of the nucleation pulse and average pressure of nucleation pulse. This takes time and the accuracy of the data depends on the subjective opinion of the user. To automate this process, a special program was created in the MATLAB environment. The interface of this program shows on the Figure 2.

The experimental data is displayed in the form of a graph, which is subsequently analyzed. Figure 3 shows a normalized scattered light flux and pressure with dependence from time. The pressure signal shows expansion with duration a few milliseconds. A significant light scattering is observed only after the nucleation pulse has occurred. Nucleation rates can be calculated by a constant-angle Mie scattering method[21].

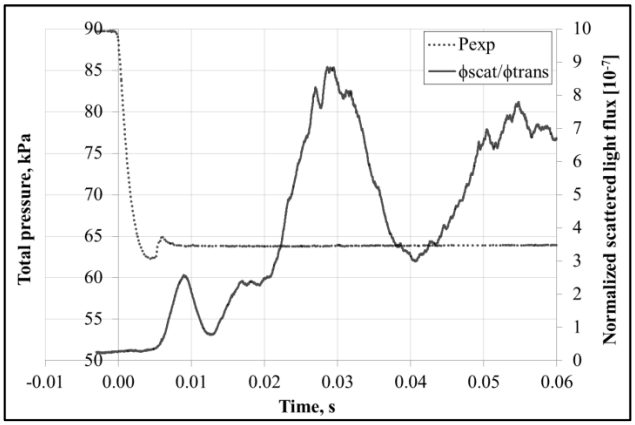

Fig. 3. Result of a typical single homogeneous nucleation experiment showing the total pressure and the light flux scattered at an angle of $\theta = 15^{\circ}$ normalized with respect to the transmitted light flux as functions of time

3 Results

An experimental study was carried out at nucleation temperatures of 220 - 260 K and initial pressures of 80 - 150 kPa, with argon and nitrogen as carrier gases. Carrier gases were chosen in this way because they represent examples of monoatomic and diatomic gases. Results of the experiments are shown on Figures 4 and 5. In the figures, the lines show isotherms.

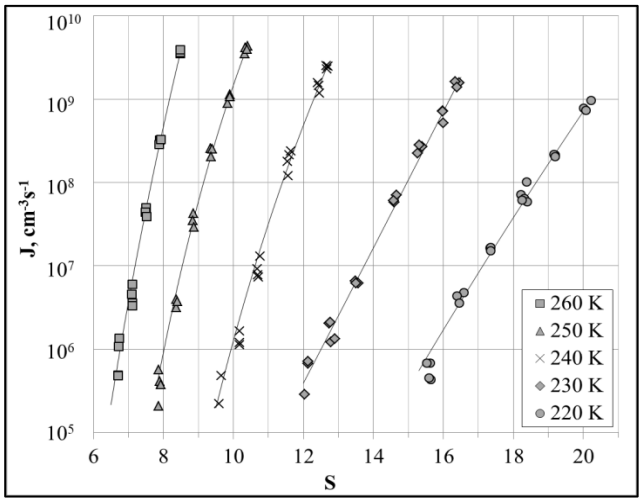

Fig. 4. Experimental data of water homogenous nucleation with argon as carrier gas.

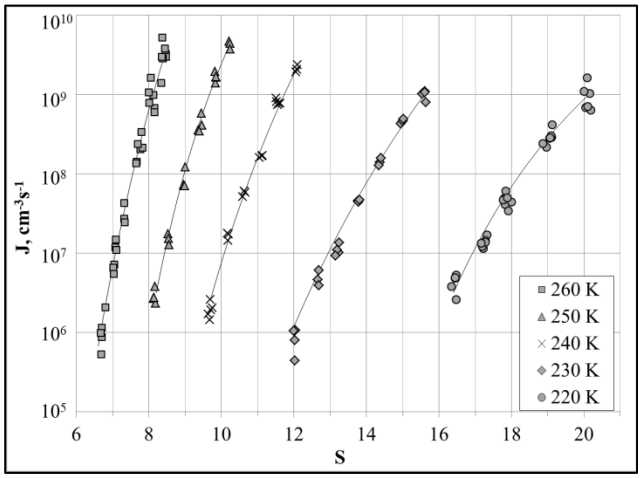

Fig. 5. Experimental data of water homogenous nucleation with nitrogen as carrier gas.

For a better visual comparison of the results, the experimental data were fitted by the equation

$$J = \exp\left(a + \frac{b}{S^c}\right), \quad (4)$$

where *a*, *b*, *c* are coefficients of equation.

Figure 6 shows a comparison of the experimental data fitted by the equation. As follows from Figure 6 on isotherms 220 – 250 K, the nucleation rate is higher at the same supersaturation with carrier gas nitrogen than with argon. This is due to the decrease in the surface tension of water because of adsorption effect of the carrier gas. As it was studied in[22] water surface tension reduction for nitrogen is grate than for argon.

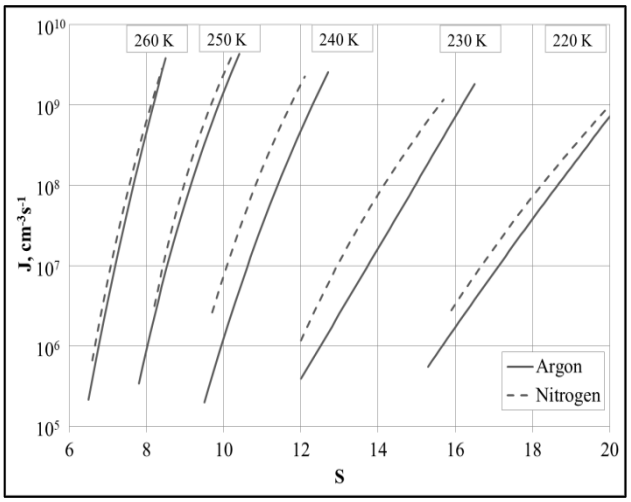

Fig. 6. Comparison of water nucleation experimental data with different carrier gases.

4 Conclusion

In the Institute of Thermomechanics, setup [2] has been completely disassembled and cleaned. Pressure transducers, laser, and the data acquisition system have been replaced with modern parts. After that, the chamber was assembled, and thoroughly tested and calibrated. To process the experimental data, a program was created in MATLAB environment, which greatly accelerated and facilitated the conduct of experiments and processing experimental data. The experiment conducted showed that the setup works properly and promising for further research. Investigation of the carrier gas influence on homogeneous water nucleation was also carried out. As a result, it was shown that due to the carrier gas adsorption effect, which leads to a decrease in the surface tension of the water [22], there is an increase in nucleation rate. This effect is more pronounced when using nitrogen as a carrier gas. Further studies of the influence of the carrier gas on the homogeneous water nucleation are planned.

Acknowledgments

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